CARBON IN PRIMITIVE METEORITES

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INTRODUCTION

No meteorites are truly primitive, in the sense of being pristine collections of interstellar grains or solar-nebular condensates. Nonetheless, some chondritic meteorites have been so little altered by secondary processing that they are commonly termed "primitive" and it is almost a definition of such chondrites that they contain significant quantities of carbon. Most of that carbon is of apparently local, i.e., solar-system, origin but a proportion that ranges from trace, in some cases, to minor, in others, is believed to be exotic, i.e., of circumstellar or interstellar origin, and it is upon such material that we focus here. However, before delving into the observational record, we must discuss briefly the nature of the meteoritic samples and the techniques used to analyse them.

Essentially all the studies performed so far have involved those meteorites known as carbonaceous chondrites. The abundant, and therefore conspicuous, carbon that gives them their name is mainly combined with hydrogen, oxygen and nitrogen to form a variety of organic compounds, discussed in more detail later. Separating the exotic carbon from the much more abundant organic matter, as well as from the lithic constituents of the meteorites, has been difficult task. The experimental approach may be briefly described as follows.

A sample of the meteorite is first demineralised using a combination of hydrofluoric and hydrochloric acids, and then oxidised, typically with dichromate and perchloric acid, to remove the organic matter. A variety of grain-size and density fractions are then prepared, which may in turn be treated with certain reagents to remove specific minerals, e.g., phosphoric acid to dissolve spinel. The residue from such treatments consists predominantly of inorganic carbides and various forms of elemental carbon. It is commonly necessary to separate such carbonaceous species further from each other, usually by means of further oxidation, including use of oxygen plasma.

Further separation is possible during subsequent analysis. An exotic origin for a carbon fraction is inferred if either the carbon itself or an element associated with it is characterised by an anomalous isotopic composition. The first step in a conventional isotopic analysis involves either pyrolysing or combusting the carbon in a gasextraction system. Such an extraction can be performed in steps, so that, in some cases, species can be separated from each other on the basis of their susceptibility to either pyrolysis or further oxidation. It is found, for example, that turbostratic carbon combusts at a lower temperature than diamond and fine-grained material before

coarse-grained. At each step, the gases evolved are collected and analysed in a mass spectrometer. Carbon itself is collected and analysed as carbon dioxide. Other gases collected and analysed are nitrogen and the noble gases, i.e., helium, neon, argon, krypton and xenon. In a few cases, hydrogen is evolved as water and is analysed isotopically after reduction to molecular hydrogen.

Recently, conventional mass spectrometry has been augmented in these studies by use of the ion microprobe. This permits isotopic, and elemental, analysis of much smaller particles than can be accommodated by conventional mass spectrometry, though at a cost of a modest reduction in precision. An ion-microprobe analysis can in principle be carried out on a particle while still embedded in a meteorite section, but in practice it is more convenient to concentrate the sample by prior demineralisation.

An anomalous isotopic composition, taken as diagnostic of an exotic origin, can be recognised in either of two ways. For elements with three or more isotopes, i.e., neon, argon, krypton and xenon, the test of anomalous behavior is whether or not the observed composition, after correction for the effects of radionuclide decay and cosmic-ray spallation, can be derived from the common terrestrial composition by mass-dependent fractionation. On a three-isotope plot, such as figure 1, mass-dependent fractionation follows a unique trajectory: deviation from that trajectory is the signature of anomalous behavior. For an element, such as carbon, nitrogen, hydrogen or helium, with only two stable isotopes, such a test is not possible. In such a case, the criterion for anomalous behavior is based on the magnitude of the observed departure from normal composition: if the deviation exceeds what can be attributed to plausible fractionation processes, the composition is regarded, at least tentatively, as anomalous.

Having determined that a carbonaceous component is isotopically anomalous and therefore probably exotic, it is obviously desirable to learn as much about its physical and chemical properties as possible. The ultimate objective of such a study is to characterise the astrophysical source of the component as fully as possible. Because the exotic components identified so far have in common a very fine grain size, the techniques available for characterisation are severely limited. Elemental analyses are restricted to the ion microprobe and various electron optical techniques, such as the electron microprobe, electron energy loss spectroscopy, analytical electron microscopy and energy-dispersive X-ray analysis using the scanning electron microscope. The same electron microscope techniques can be used to image the carbonaceous particles, and high-resolution transmission electron microscopy can also yield crystallographic information. Further crystal-structural data can be obtained by means of electron diffraction and laser Raman microprobe analysis. Because these different analyses are invariably performed on different splits of the same sample, it is not always possible to establish a rigorous one-to-one correspondence between, say, the particle imaged in the electron microscope and the noble-gas component measured in the mass spectrometer. Many of the associations described below are therefore inferential.

Before discussing the anomalies observed so far, it is desirable to expand somewhat on what we mean by normal. As noted above, this is largely based on terrestrial values but in some cases this choice of standard warrants elaboration.

WHAT IS NORMAL?

Because we are interested in identifying material that originated outside the solar system, we first need to establish the average solar system composition. Many tabulations of such "cosmic" abundances have been published, and for the great majority of the elements there is good evidence that those abundances are very reliable guides to the composition of the early solar system (e.g., ref. 1). However, for the five elements of most concern to us here, i.e., hydrogen, carbon, nitrogen, neon and xenon, a variety of factors make it desirable to discuss briefly their isotopic compositions in the early solar system.

Hydrogen

The terrestrial value of D/H is 1.55 x 10⁻⁴. However, this value undoubtedly reflects substantial mass-dependent fractionation during accretion and early evolution of the earth. Because D is destroyed by nuclear reactions in the Sun, we cannot be guided by solar spectroscopy or solar-wind analysis. Our best estimates for the protosolar value come from observations of the present interstellar medium (ref. 2), spectroscopy of the atmospheres of the giant planets (ref. 3) and reconstructions based on the present ratio of 3He to 4He in the sun, much of the 3He having been derived from primordial D (ref. 4). These approaches all converge on a value for D/H of about 2 x 10⁻⁵ for the hydrogen from which the solar system was made (ref. 5), though there is good evidence that the H isotopes were inhomogeneously distributed in the solar nebula, so that a unique solar-system ratio may have no physical significance (ref. 3)

Carbon

The terrestrial value for $^{12}\text{C}/^{13}\text{C}$ is about 89, and a closely similar value is also observed in the solar wind (ref. 6) so that we are probably justified in taking it as characteristic of the solar system as a whole. Considerable variability in this ratio is observed for the interstellar medium but the data are believed to be consistent with a value around 90 at 4.5Gyr ago (ref. 7).

Nitrogen

A value of 276.8 characterises the $^{14}\rm{N/^{15}N}$ ratio in the terrestrial atmosphere. The solar ratio is not uniquely determined, values ranging from 247 to 360 having been inferred for the solar wind at different epochs (ref. 8). Bulk meteorite values range from 140 to

over 300, but because nitrogen is grossly depleted in most meteorites relative to its cosmic abundance, even bulk values could be significantly affected either by isotopic fractionation prior to, during or after agglomeration of the meteorite or by the presence of anomalous components. The composition of the interstellar medium is not helpful in this regard, and values for the giant planets are imprecise at this time, though the Galileo mission to Jupiter will greatly improve that situation. Nonetheless, as for hydrogen, it is quite plausible that there was no unique, solar-system-wide value for $^{14}\text{N}/^{15}\text{N}$, i.e., that primordial solar-system nitrogen consisted of more than one isotopic component, presumably of nucleogenetic origin (ref. 9).

Neon

The isotopic composition of neon in the solar wind is precisely constrained by measurements made on the lunar surface (ref. 10). On a three-isotope plot, figure 2, all local, i.e., solar-system, values fall within the triangle bounded by "solar wind", "spallation" (resulting from cosmic-ray interactions) and a common meteoritic value, termed neon-A, of uncertain origin. Curiously, a composition equivalent to that of neon-A has been observed in solar flares (ref. 11). Because of the uncertain relationship between solar-wind neon and neon-A, it is not clear whether the solar nebula was homogeneous with respect to the neon isotopes. However, as will be seen, the magnitude of the observed anomaly vastly outweighs that uncertainty.

Xenon

Defining a unique proto-solar-system value for the xenon isotopes, if in fact such a value existed, is not straightforward (ref. 12), but fortunately the plethora of xenon isotopes and the variety of astrophysical sites in which they are made have made anomalous xenon components fairly easy to recognise.

ROSTER OF ANOMALOUS CARBON COMPONENTS

The exotic carbon components that have been recognised so far in meteorites, based on the anomalous isotopic composition of one or more elements, are listed in table 1, together with a summary of their properties. The reader is reminded that the association of such properties with specific phases is in some cases inferential and may therefore be subject to change in light of future results. In what follows, we discuss each component in some detail. The study of some we shall focus mainly on the results of the most recent studies. For convenience, we shall employ the nomenclature advanced by Anders and his colleagues (e.g., ref. 13), as in table 1. Although the distribution of these components among different types of meteorites has not been fully mapped out, it seems likely that all of those listed in table 1 occur to a greater or less extent in all carbonaceous chond-

Name	Сα	Св	C &	Q	Сε	Organic
Mineralogy		SiC	Diamond	Diamond?	SiC	
Anomaly	Ne-E(L)	Xe-S	Xe-HL	Planetary	Ne-E(H)	Hydrogen
Enrichment	²² Ne	¹³⁰ Xe	124,136 Xe	Heavy	^{2 2} Ne	D
Pyrolysis T ^O C	700	1400	1000	variable	1200	300-1300
Combustion T ^O C	600	1000	500	500	1000	250 - 450
12C/13C	66.5	36 - 56	92.5	92.5?	<12.7	85.3-90.8
14N/15N	< 221	395-462	413	?	< 358	252-278
D/H x 10 ⁻⁵			∿20.3	?		?-62.4
	5	2	400	(as C&?)	7	2-3wt%
Abundance (µg/g) Grain size (µm)	1-10	0.03-0.2	0.0026	(as C&?)	0.03-1	
Probable source	nova	red giant	C: red gian	it (as C&?)	nova?	molecular cloud(s)
			Xe: supernov	ra		nebula?
						asteroid?

TABLE 1. Summary of the apparently exotic carbonaceous components identified so far in primitive meteorites. Based largely on ref. 13.

rites. Most studies have focused on Orgueil (type CI), Murchison (CM2) and Allende (CV3).

Carbon Alpha

This component was recognised by the isotopically anomalous neon that it contains. This neon, termed neon-E, is essentially pure 22 Ne (ref. 14), as can be seen in figure 2. Careful scrutiny of that figure will also reveal that neon-E is released in two distinct temperature ranges: one around 700°C and one around 1000°C. The neon released in these ranges is termed neon-E(L) and neon-E(H), respectively. Carbon alpha is the host phase of the neon-E(L). Both neon-E components are believed to have been produced by decay of 22 Na, which has a half life of 2.6yr. Because of the shortness of that half life, decay must have taken place in the atmosphere of the star in which the taken place in the atmosphere of the star in which the towards that star having been a nova (ref. 13). Selective filtration indicates that carbon alpha has a grain size between 1 and 10 micrometers (ref. 13), thereby supplying a provisional measure of the grain growth achieved in nova outflows. Carbon alpha has not been well characterised but its low density, below 2.4g/cm³, hints at a form of elemental carbon.

Carbon Beta

The signature of carbon beta is a xenon component that matches with remarkable accuracy the composition predicted to be produced by the s-process (nucleosynthesis that proceeds by the capture of neutrons on a time scale that is long compared with the half lives of the species being produced). This match between theory and observation is illustrated in figure 3 (ref. 15). Physically, carbon beta has been identified as silicon carbide with a grain size in the range 0.03 to

0.2 micrometers (ref. 16). The silicon and the carbon are both substantially enriched in the heavy isotopes: the $^{30}\text{Si}/^{28}\text{Si}$ ratio, not given in table 1, is up to 5% higher than the terrestrial value (ref. 17). Nitrogen, whose physical state in carbon beta is unknown, is heavily depleted in ^{15}N ($^{14}\text{N}/^{15}\text{N}$ as high as 462). An origin for carbon beta in a red giant star has been proposed (ref. 13) n, whose physical state in carbon beta is unknown, is ted in ^{15}N ($^{14}N/^{15}N$ as high as 462). An origin for carbon giant star has been proposed (ref. 13).

Carbon Delta

aceous regions and xenon-H in neutrexplosively ejected from the diamond structure by substitution for carbon. tly developed into a supernova, creating regions and xenon-H in neutron-rich zones. T 12c/13c ratio is 92.5, remarkably (13). One model to explain the prope condensation of the diamond in the attime freshly synthesised nitrogen not aggregates measuring up to about 0.5 micrometers in diameter, figure (ref. 21). As can be seen in figure 4, the anomalous xenon consists two components, one enriched in the heavy isotopes (H), the other light (L)(ref. 13. These com-ponents are closely coupled and it accompanies 12c/13c rati The presence of this component is also recognised by anomalous (enon, as shown in figure 4. The host phase of this xenon appears to be diamond with a grain size of only 10s of angstroms (ref. 18). Perhaps because of this fine grain size with the consequent large proportion of surficial carbon, as much as 25% of carbon delta appears to behave as amorphous material (ref. 19). At least in the acidesistant residues studied so far, the tiny crystallites occur as aggregates measuring up to about 0.5 micrometers in diameter, figure 5 alternative proved possible to separate them experimentally. The nitrogen that ompanies xenon-HL is highly enriched in 14N, table 1, but its / 13C ratio is 92.5, remarkably close to the "local" value (ref. one model to explain the properties of carbon delta postulates e freshly synthesised nitrogen was also incorporated into the mond structure by substitution for carbon for the corporated into the veloped into a supernova, creating xenon-L in neutron-poors and xenon-H in neutron-rich zones. The resulting mixture was ively ejected from the supernova, overtook the relatively slowly diamond grains, and became implanted in them (ref. 13 and 20). ernative model (ref. 19) invokes melting of interstellar carbonfrom the by supernova shock waves followed by nucleation o transiently high-pressure liquid. The red giant subsequen-kenon-L in neutron-poor callites occur as diameter, figure 5 xenon consists of ín

Q

delta characterised by the presence of a noble-gas population that seem be prevalent on planetary objects in the solar system, figure to the ver, these noble gases are closely associated in nature vecon-HL, though readily separated from it in the laboratory, so ref. 21) that Q-gas is trapped in pores at the surfaces of callelta grains. This trapping is postulated to have taken place in solar nebula and to have been accompanied by substantial fracticion, resulting in the observed systematic compositional differenteem Q-gas and solar gas (ref. 21 and 22). strict sense, the component termed Q is not anomalous; has been suggested surfaces of carbon taken place in the ystem, figure 6. I in nature with boratory, so that s been suggested fractiona differenc figure to 6.

Carbon Epsilon

- 8 perature. Th icon carbide As noted earlier, neon-E has a bimodal release as a functure. The host of the higher-temperature fraction is function of another

respects features isotope of nitrogen, carbon epsilon is characterised by much greater enrichments than carbon beta (ref. 17). In addition, the siliconisotopic data for carbon epsilon require a minimum of three nucleogenetically distinct components (ref. 17). Although the origins of those silicon components are not yet clear, the other isotopic properties of carbon epsilon indicate a nova origin, as for carbon beta. tures to identify it as a separate phase. Thus, although both are iched in the heavy isotopes of silicon and carbon and the light cts, it resembles carbon beta but possesses enough distinguishing

Organic Matter

It seems likely that more than one process contributed to the currently present in the meteorites. Isotopically, the organism undane, in fact it is not inconceivable that it major source of and inconceivable that it is not inconceivable that it is major source of and inconceivable that it is major source of an inconceivable that it is major as mundane, in fact it is not inconceivable that it major source of carbon for the terrestrial planets. organically combined nitrogen, although it exhibits topic variability, does not fall outside the range fractionation. about 3wt% and 2wt%, respectively (ref. 23). At the molecular levels this organic matter consists of a complex mixture of a wide variety organic compounds (ref. 24). The origin of this material is disputed an entire compounds (ref. 24). argument. tainty about ractionation primitive chondrites, at only trace levels. By on is a major component in both CI and CM chondr The components n by possible solar-system processes, though the the protosolar $^{14}{\rm N}/^{15}{\rm N}$ ratio reduces the force discussed respectively 80 far have h CI and CM chondrites, (ref. 23). At the mol Isotopically, the organic ceivable that it represent restrict been exhibits significant the range of present, though the molecular contrast, represented Similarly, amounting even plausible disputed: o f melang organic unceræ

isotopic hydrogen molecule reactions taking place at very low translational (ref. 5, 27 and 28). Such reactions are believed to be rethe extremely large deuterium enrichments observed in gas cules of dense interstellar molecular clouds (ref. 29), leading conclusion that some fraction of the organic hydrogen in mete originated in such interstellar clouds (ref. 30). Exactly in what this anomalous hydrogen traversed interstellar space and lentered the solar nebula and became combined with apparently enriched carbon to form organic molecules represent problems understood. onsistent f meteoritic and 26). These not solar-nebular or planetesimal processes, attributed to the isotopic fractionation fully od. Preservation of the interstellar (
of abundant low-deuterium hydrogen in
a serious constraint on model composition of its hydrogen. T isotopes g., ref. 24). lly understood at this time. However, existing data are with all meteoritic organic molecules being enormously deuterium among the enrichments meteoritic organic molecules being enormousl with respect to protosolar hydrogen (e.g., ref matter its hydrogen. The detailed different organic constituents to exceed the fractionating interstellar deuterium catalysed fractionations its believed to be responsive mole gas-phase exotic 30). Exactly in what form tellar space and how it attribute the so that the associated character is distribution of is comp that solar nebula reactions apparently capacity of they are com-ated with ionce and how it parently local care not fully n signature in temperature complex to th conany S

CONCLUSIONS

The components discussed above by no means exhaust the list of carbonaceous phases present in meteorites but include all those for which an extrasolar signature has been reliably observed. Two other, probably local, carbonaceous components of meteorites deserve mention, however. Inorganic carbonates are common constituents of CI and CM carbonaceous chondrites and it has been suggested that these might also be of interstellar origin (ref. 31). However, their compositions, morphology and petrology show clearly that they were formed by aqueous activity on asteroidal parent bodies (ref. 32).

In addition, poorly graphitised (or turbostratic) carbon (PGC) occurs in many ordinary and carbonaceous chondrites together, in most cases, with amorphous carbon. In ordinary chondrites, such material is also commonly associated with fine-grained metallic iron (ref. 33). Well-ordered graphite appears to be very rare in these primitive meteorites. Textural relations suggest that many, if not all, chondritic occurrences of PGC resulted from dehydrogenation of kerogen-like organic matter (ref. 33 and 34). Such kerogen-like material is the dominant constituent of the organic matter in carbonaceous chondrites (ref. 24).

Nuth (ref. 35) has pointed out that the extreme scarcity of graphite in primitive meteorites that contain measurable amounts of interstellar carbon in other, less refractory forms, suggests strongly that graphite is similarly rare in the interstellar medium and that spectral features that have commonly been attributed to graphite are probably due to other species. He also inferred that grain lifetimes in the interstellar medium are considerably longer than the $10^8 \, \mathrm{yr}$ of most generally accepted models.

Clearly, the study of exotic carbon preserved in meteorites has been informative about sites of nucleosynthesis, processes of nucleation and growth of grains in stellar outflows, grain survival in the interstellar medium, and many other topics of astrophysical significance. Much more work, particularly of an interdisciplinary nature remains to be done, however.

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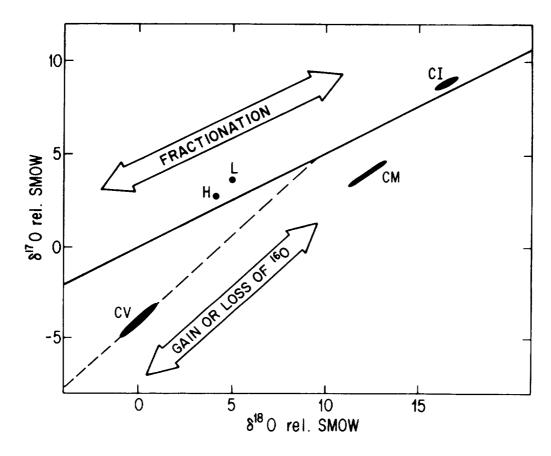


Figure 1.– Three-isotope plot for the oxygen isotopic system. The ratio ¹⁷0/¹⁶0 is plotted against ¹⁸0/¹⁶0 for several groups of chondrites. Ratios are expressed as deviations (delta units) in parts per 10³ relative to the ratios for Standard Mean Ocean Water. On such a plot, mass-dependent fractionation defines a line with a slope close to 0.5, as shown by the solid line, which is the locus of all data for terrestrial samples. Deviations from that line, as exhibited by the chondritic data, cannot be explained by such fractionation but apparently require addition or subtraction of one or more anomalous components, such as the hypothetical pure ¹⁶0 one illustrated. Data from the laboratory of R. N. Clayton.

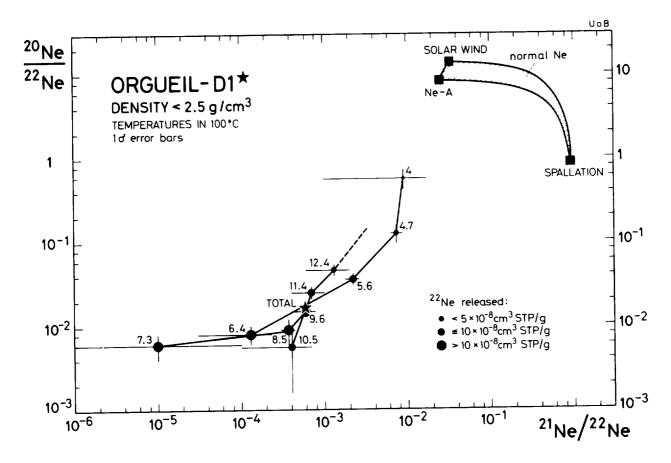


Figure 2.— Three-isotope plot for Ne extracted from a low-density fraction separated from the Orgueil carbonaceous chondrite. Note the logarithmic scale and the enormous enrichment of the D1 fraction in ²²Ne, which amounts to more than 98% of the Ne released. Data from the laboratory of P. Eberhardt. Figure from Anders (1988).

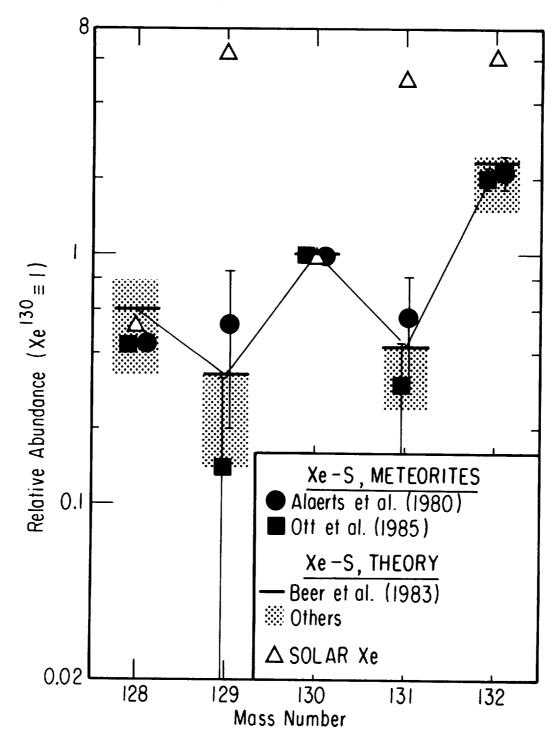


Figure 3.— The isotopic composition of the meteoritic Xe-S component, compared with the theoretical yield of s-process nucleosynthesis and with solar Xe. The match with s-process theory is excellent. Figure from Anders (1988).

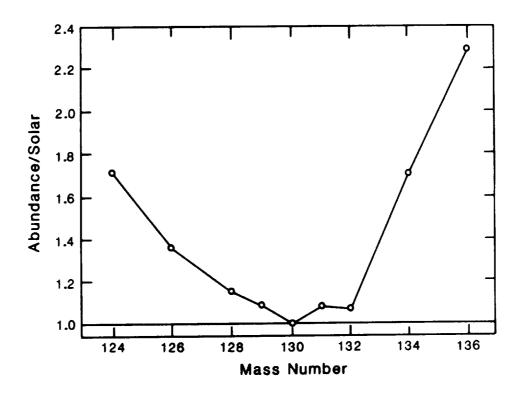


Figure 4.– The isotopic composition of a meteoritic Xe component enriched in both heavy and light isotopes and therefore known as Xe-HL.

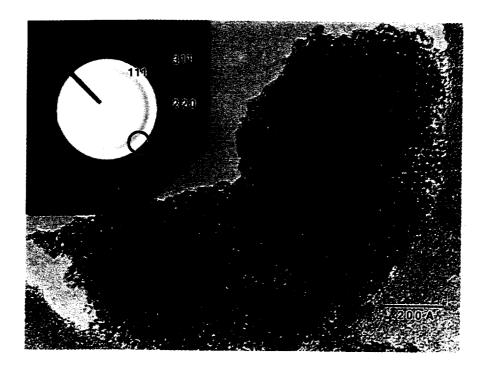




Figure 5.— Bright-field (upper) and dark-field (lower) images of diamond in a C delta residue from the Allende meteorite. Inset diffraction pattern can be indexed as diamond. Dark-field image obtained using a portion of the (111) powder ring indicated in the diffraction pattern. Bright irregular regions in dark-field micrograph correspond to 5-70Å crystallites of diamond which are in proper orientation for diffraction. Courtesy of David Blake.

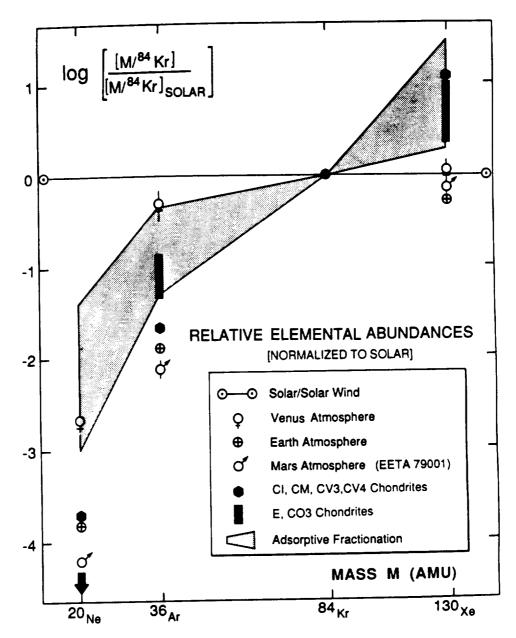


Figure 6.– Noble-gas abundance ratios (normalized to Kr) in atmospheres of terrestrial planets and in volatile-rich meteorites. Despite differences in detail, those inventories, loosely categorized as "planetary gas," have in common an enrichment in the heavy gases relative to solar composition. In primitive chondrites, planetary gas is sometimes termed "Q gas." Figure from Hunten, Pepin and Owen (ref. 36).

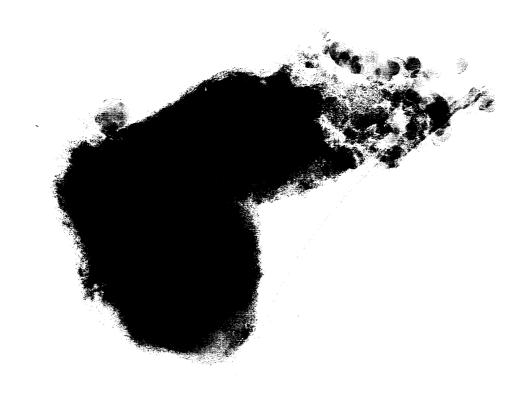


Figure 7.– Transmission electron micrograph of a SiC particle, identified as Carbon Epsilon, from the Murray carbonaceous chondrite. The scale bar measures 2000Å. Attached to the SiC are some fine-grained diamonds and round blobs of a Si-oxide phase. Micrograph courtesy of Thomas Bernatowicz and Ernst Zinner.